# Computational Approaches to the Chemistry of Nanopores

Sandia National Laboratories

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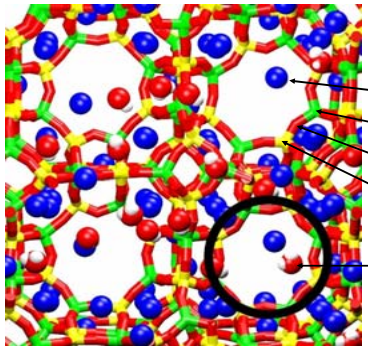
## The Problem

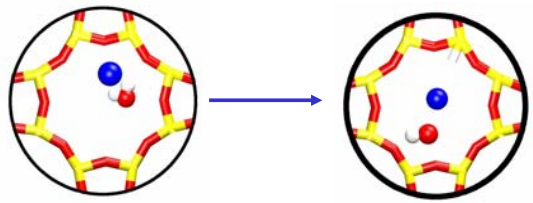
**Modeling local chemistry in the presence of collective phenomena is a critical step toward scientific understanding of complex nanoporous materials**

Zeolite-4A collective phenomena involve 672+ atoms.

These length and time scales demand classical treatment.



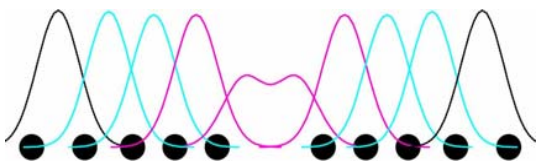

Zeolite-4A chemical specificity involves a few atoms, but requires the accuracy and flexibility of *ab initio* quantum methods.



## IDEA: Embed Density Functional Theory in a Classical Background

**Efficient embedding: A localized view of electronic structure based on "Generalized Wannier Functions"**

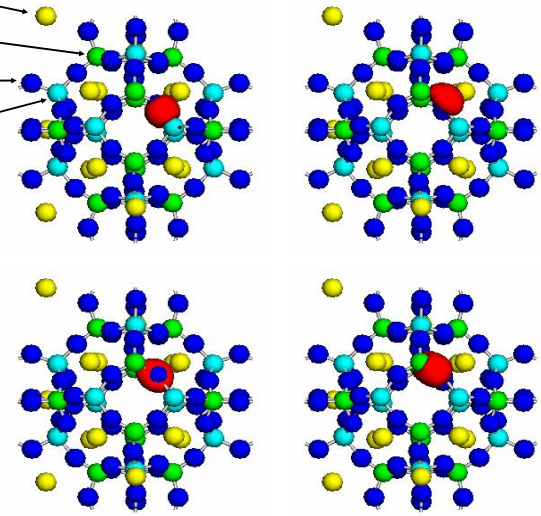
- Linearly independent localized functions that span occupied subspace of the Kohn-Sham Hamiltonian
- The density matrix and total energy are easily evaluated
- Optimize red, approximate blue, and ignore black



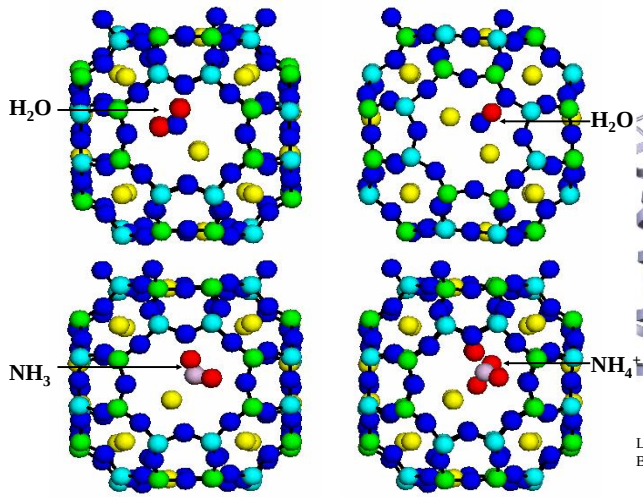
**Two methods of solving for GWF have been implemented**

- Green's Function Approach
- Grassmann Conjugate Gradient Approach

**The Generalized Wannier Functions from the Green's Function approach are naturally localized**



## Results: $H_2O$, $NH_3$, and $NH_4^+$ in Zeolite-4A




## Funding

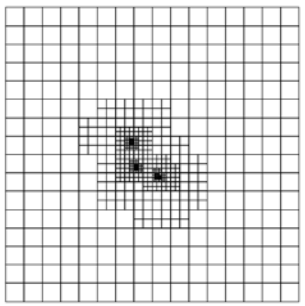
Sandia Laboratory Directed Research and Development

New Proposal to DOE joint BES/ASCR Computational Nanoscience Call: **Investigating the Chemistry of Nanopores Using a Localized Hierarchical Basis**

Our Team: Normand Modine, Pavel Bochev, Mark Sears, Richard Lehoucq, Susan Rempe, Kevin Leung, Michael Chandross, Tomas Arias (Cornell), Chris Anderson (UCLA)

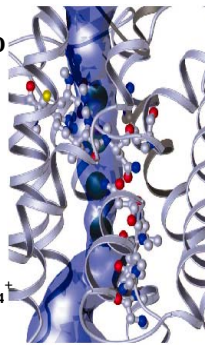
The Idea: Work with the mathematicians to address efficiently the range of scales involved in the chemistry of nanopores

**A Hierarchical Basis is key to an efficient treatment of inhomogeneous length scales**



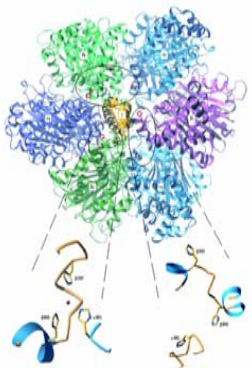
**Study remarkable selectivity and switching of natural and artificial nanoporous systems**

**Natural Biological Channels**



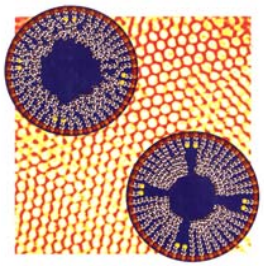
Law and Sansom, Current Biology **12**, R250 (2002).

**Bio-engineered switchable proteins**



Liu et al., Nature Maters. **1**, 173 (2002).

**Functionalized Nanocomposites**



From Jun Liu, Sandia